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PROGRESS REPORT NO. 30-20

**THE THERMAL EXPANSION OF SYNTHETIC GRAPHITES AT
TEMPERATURE INTERVALS BETWEEN 80 AND 2000°F**

ROBERT D. ALLEN

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ABSTRACT

The mean linear and cubical coefficients of thermal expansion of eight commercial lots of graphite were determined for temperature intervals between 80 and 2000°F. The linear thermal expansion was measured with an automatic recording dilatometer using a rod-shaped specimen 2 in. long and ¼ in. across. The specimen was heated in an atmosphere of helium. The results are in good agreement with those of Currie, Hamister, and MacPherson. The mean linear coefficient was found to increase with temperature. For the lots studied, the mean linear coefficients from 80 to 2000°F were 1.50 to $2.34 \times 10^{-6}/^{\circ}\text{F}$ parallel and 2.26 to $3.45 \times 10^{-6}/^{\circ}\text{F}$ perpendicular to the grain and were found to vary linearly with the electrical resistivity measured at 32°F.

I. INTRODUCTION

In high-temperature design applications it is important to know the mean coefficients of linear thermal expansion of synthetic graphite over temperature intervals from room temperature to the region of 5000°F. This study, the first of a series in a high-temperature thermal expansion program, provides information about eight commercial grades of molded and extruded graphite at temperatures between 80 and 2000°F.

Earlier investigations¹ such as those by Hidnert (Ref. 1) and Burdick, Zweig, and Moreland (Ref. 2) indicate the

¹See Bibliography.

following conclusions: (1) the coefficient of expansion of graphite usually undergoes a gradual increase with increase in temperature; (2) specimens cut perpendicular to the grain direction have greater expansions than specimens cut parallel to the grain direction.

Currie, Hamister, and MacPherson (Ref. 3) have found that the mean linear coefficient of thermal expansion between room temperature and any final temperature can be obtained from the coefficient for the interval room temperature to 212°F by adding to it an appropriate number.

II. DESCRIPTION OF GRAPHITES TESTED

The nominal characteristics of the graphites tested are presented in Table 1. These grades of graphite were selected because they represent a variety of manufacturing conditions and because the high-temperature tensile

and creep properties have previously been described (Ref. 4). Measured specific gravity data were calculated from the weights and dimensions of thermal expansion specimens before testing.

Table 1. Nominal Characteristics and Measured Densities of Graphites Tested

(All data except measured specific gravity supplied by manufacturers)

| Block ^a | Commercial Grade Designation, Manufacturer | Method of Forming | Size of Block ^b , in. | Maximum Filler Particle Size, in. | Starting Materials ^c | Impregnation | Graphitizing Temperature, °F | Nominal Specific Gravity | Measured Specific Gravity |
|----------------------|--|-------------------|---|-----------------------------------|---------------------------------|--------------|------------------------------|--------------------------|---------------------------|
| 0104 0105 0107 | 3499; Speer Carbon Co. | molded | 12 × 12 × 2 | 0.005 | coke, pitch | pitch | above 4100 | 1.68 | 1.54–1.61 |
| 0201 | 942S; Speer Carbon Co. | extruded | 2½ × 17 × 24 | 0.033 | coke, pitch | pitch | above 4100 | 1.65 | 1.62–1.67 |
| 0301 | AGR; National Carbon Co. | extruded | 16 × 16 × 60 | 0.250 | coke, pitch | none | 5000 | 1.51 | 1.54–1.55 |
| 0401 | H3LM; Great Lakes Carbon Co. | extruded | 12 D × 72 | 0.06 | coke, pitch | pitch | 5075 | 1.70 | 1.74–1.75 |
| 0501 | 3499; Speer Carbon Co. | molded | 12 × 12 × 2 | 0.005 | coke, pitch | pitch | above 4100 | 1.68 | 1.56 |
| 0601 | EH; Speer Carbon Co. | molded | 12 × 6 × 2 | 0.005 | graphite flour, pitch | pitch | above 4100 | 1.65 | 1.64 |
| 0701 | 896G; Speer Carbon Co. | extruded | 16 × 16 × 2 cut from 16 × 16 × 72 | 0.033 | coke, pitch | none | above 4100 | 1.54 | 1.50–1.54 |
| 0801 | ATJ; National Carbon Co. | molded | 20 × 24 × 6 | 0.005 | coke, pitch | pitch | 5000 | 1.77 | 1.69–1.75 |
| 1001 1002 | NT0005; National Carbon Co. ^d | molded | — | — | — | — | — | not supplied | 1.72–1.78 |
| 1101 | AGX; National Carbon Co. | extruded | ¾ D × 12 | — | coke pitch | — | 5000 | — | — |

^aBlocks from the same commercial grade (excepting block 0501) have the same first two digits.

^bDimension on right is parallel to the molding or extrusion direction.

^cCoke is petroleum coke throughout; pitch is coal tar pitch throughout.

^dExperimental grade; baked under pressure, not graphitized. Other manufacturing information not disclosed.

III. EXPERIMENTAL PROCEDURE

In molded graphite the grain direction tends toward perpendicularity to the direction of molding pressure, whereas in extruded graphite the grain direction tends to be parallel to the extrusion direction. Stated in another way, graphite crystallites tend to be oriented with their planes perpendicular to a molding direction or parallel to an extrusion axis. The orientations of the specimens were selected to make possible the determination of thermal expansion in three mutually perpendicular directions, one of which is parallel to the direction of molding pressure (perpendicular to the grain direction) or parallel to the extrusion axis (parallel to the grain direction). The letters *M*, *N*, and *P* for molded graphite and *E*, *F*, and *G* for extruded graphite specify orientations of specimens within parent blocks as shown in Fig. 1.

An automatic recording silica dilatometer, described by Duwez and Martens (Ref. 5), has been used in this work. The changes in length and temperature of the test specimen are recorded as functions of time on separate charts. Although this equipment was originally designed for use with materials with higher coefficients of expansion than those reported for graphite, it was found that satisfactory thermal expansion curves were obtained by using the 400:1 magnification.

Rod-shaped graphite specimens $\frac{3}{16}$ to $\frac{1}{4}$ in. in diameter and about 2 in. in length were used. Lengths and weights of the graphite specimens were determined before and after each run. No significant change in length was found in any sample, even, in one instance, after 15 repetitive runs. Nevertheless, a weight loss of the order of 0.1% was usually detected after a single run; the weight loss after a succeeding run was usually appreciably less than 0.1% and sometimes almost negligible.

Each specimen was placed at the bottom of the silica tube and centered with respect to the tube walls by means of a thick-walled graphite cylinder whose inside diameter was several thousandths of an inch larger than the diameter of the test specimen; the graphite cylinder also served to prevent the specimen from substantial tilting during or after insertion. The temperature of each specimen was measured with a chromel-alumel thermocouple whose junction was placed at the top of the graphite cylinder near the midpoint of the sample axis. The silica tube was flushed with helium throughout every run. No

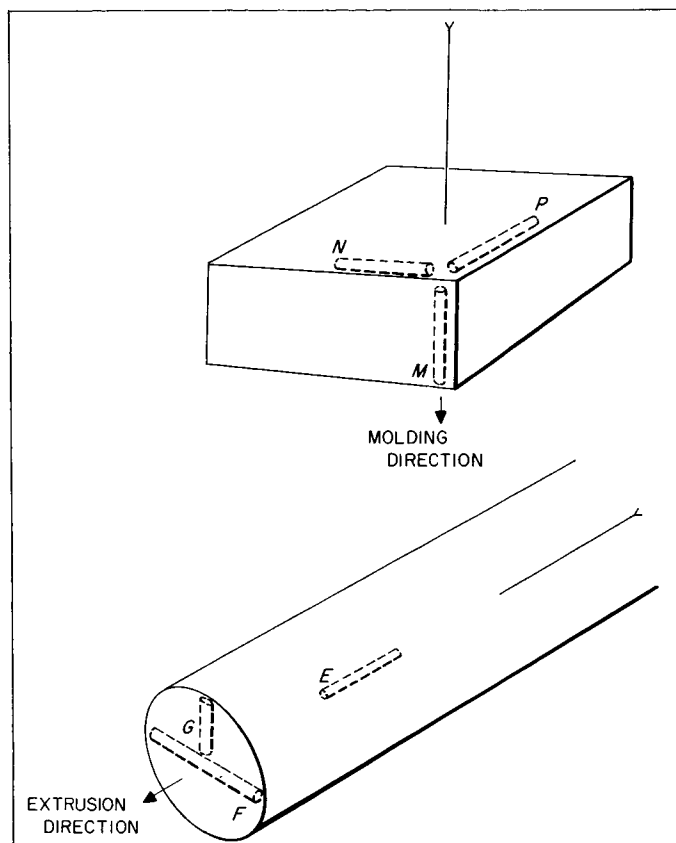


Fig. 1. Orientations of Test Specimens with Respect to Molding and Extrusion Direction

visual evidence of reaction between graphite and silica was observed. Corrections for the expansion of the silica tube parallel to the sample were made. For these corrections it was assumed that the coefficient of linear thermal expansion of fused quartz was $0.30 \times 10^{-6}/^{\circ}\text{F}$. This value was taken as an average over the temperature range studied as reported by Souder and Hidnert (Ref. 6).

The dilatometer was operated according to the following program: heat rate, 540°F per hr from room temperature to 2000°F ; soak time, $\frac{1}{2}$ hr at 2000°F ; cool rate, 540°F per hr from 2000°F to about 600°F . (The cool rate below about 600°F was somewhat less because of decrease in radiative heat loss.)

Electrical resistivities were measured with a precision Kelvin bridge. The same graphite specimens used in the thermal expansion apparatus were tested. Voltage probes

were separated by 1 in. as shown in Fig. 2. Measurements were carried out at 32°F by immersing specimens in distilled water at this temperature. Each electrical resistivity reported represents an average of 30 readings. The polar-

ity was reversed after every reading; the position of the specimen was changed after the tenth and twentieth readings. The standard deviation of the mean of 30 was less than 0.004×10^{-3} ohm cm.

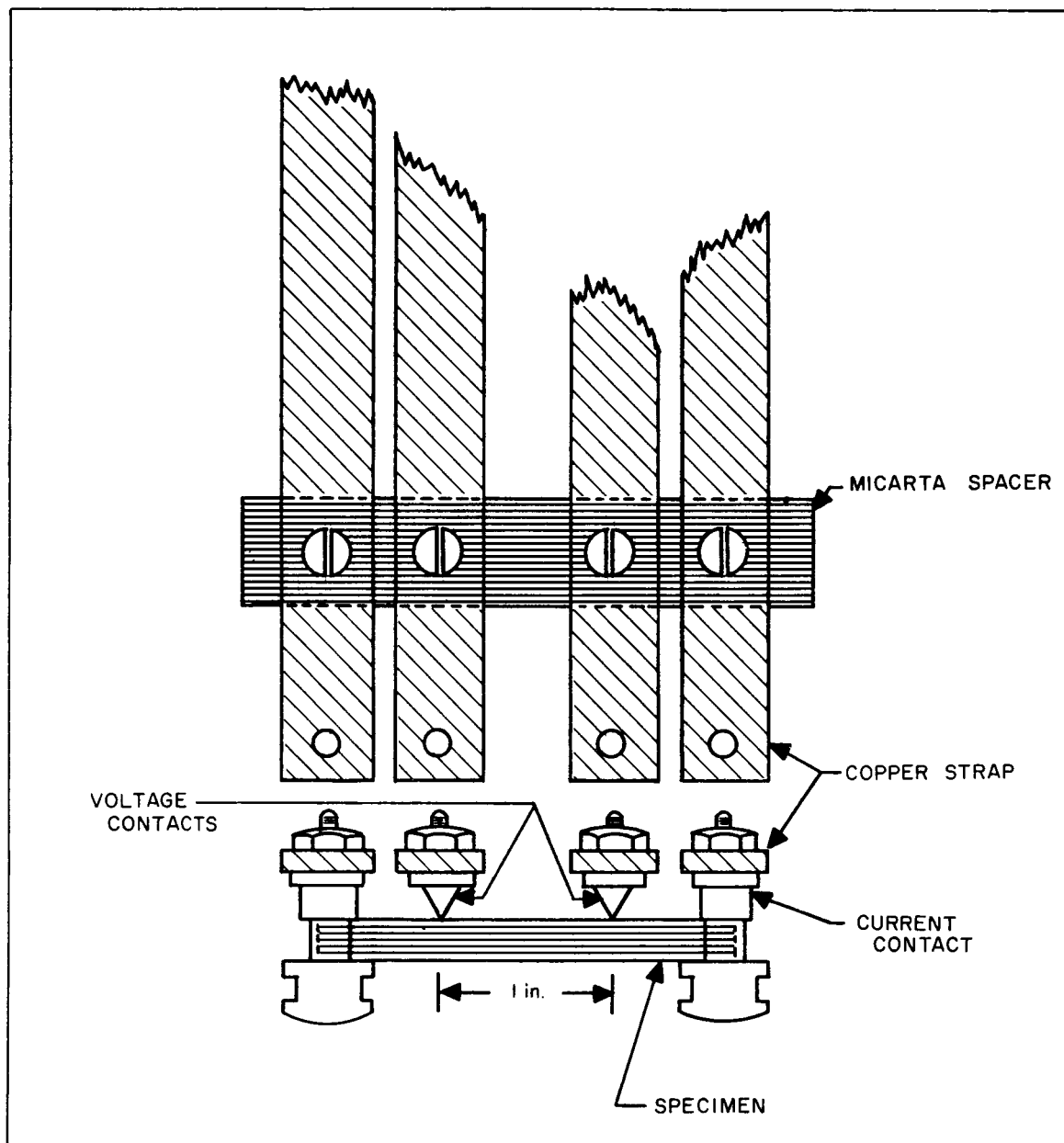


Fig. 2. Specimen Holder for Measuring Electrical Resistivity of Graphite

IV. RESULTS

A. Experimental Errors

Several standard materials were run in the dilatometer to determine its absolute accuracy. The deviations from published data are considerably less than the scatter shown by a series of runs for all temperature ranges considered except the interval from 80 to 200°F. A substantial negative deviation found in the 80 to 200°F interval is probably due to temperature lag of the test sample behind the temperature of the thermocouple in the early portion of a run. In the temperature intervals above 200°F this effect is much less pronounced, becoming negligible in the interval from 80 to 600°F and in the succeeding larger intervals. These findings are shown in Table 2, which contains the comparative data for "A" nickel and Armco iron. Comparable results were obtained for samples of electrolytic copper and AISI 303 stainless steel.

The repeatability of the equipment was examined by carrying out a series of test runs on three cylindrical sam-

ples of block 1101 extruded graphite rod, 0.1875 in. across and 2 in. long, whose lengths were parallel to the extrusion axis.

The average coefficients of thermal expansion and scatters for the several temperature intervals considered are given in Table 3. It is apparent from Table 3 that the ratio of the scatter to the average coefficient would usually be least for the 80 to 2000°F interval. Comparisons of data have been carried out in an effort to find significant differences between samples in this range.

Estimated standard deviations s were calculated for the three samples in Table 3 (80 to 2000°F interval) using the equation $s = [\sum (\bar{x} - x)^2 / (n - 1)]^{1/2}$. The results were as follows:

Sample 1: $0.102 \times 10^{-6} / ^\circ\text{F}$

Sample 2: $0.116 \times 10^{-6} / ^\circ\text{F}$

Sample 3: $0.103 \times 10^{-6} / ^\circ\text{F}$

Table 2. Linear Thermal Expansions of Standard Metals

| "A" Nickel | | | |
|--------------------------|--|--|---------------------------------------|
| Temperature Range, °F | This Investigation (average of 10 runs), % | International Nickel Co. ^a , % | Nix and MacNair ^b , % |
| 80-200 | 0.054 | 0.07 | 0.095 |
| 80-400 | 0.22 | 0.23 | 0.245 |
| 80-600 | 0.40 | 0.41 | 0.42 |
| 80-800 | 0.59 | 0.60 | 0.60 |
| 80-1000 | 0.78 | 0.79 | — |
| 80-1200 | 0.98 | 0.98 | — |
| 80-1400 | 1.20 | 1.17 | — |
| Armco Iron | | | |
| Temperature Range, °F | This Investigation, % | Nix and MacNair ^b , % | Armco Steel Corp. ^c , % |
| 80-200 | 0.070 | 0.080 | 0.0790 |
| 80-400 | 0.220 | 0.228 | 0.2275 |
| 80-600 | 0.390 | 0.395 | 0.3980 |
| 80-800 | 0.575 | 0.575 | — |
| 80-1000 | 0.767 | 0.760 | — |
| 80-1200 | 0.955 | 0.940 | — |

^aTechnical Bulletin T-15, 7th ed.
^bNix, F. C., and MacNair, D., "The Thermal Expansion of Pure Metals: Copper, Gold, Aluminum, Nickel, and Iron," *Physical Review*, 60: 597-605, 1941.
^cWritten communication.

Table 3. Average Coefficients of Thermal Expansion and Ranges of Variation for Series of Tests on Block 1101

(All coefficients determined parallel to the grain direction)

| Temperature Range, 80°F to | Sample 1 (6 runs) | | Sample 2 (6 runs) | | Sample 3 (15 runs) | |
|-------------------------------|---|---|---|---|---|---|
| | Coefficient $\times 10^6/^{\circ}\text{F}$ | Scatter $\times 10^6/^{\circ}\text{F}$ | Coefficient $\times 10^6/^{\circ}\text{F}$ | Scatter $\times 10^6/^{\circ}\text{F}$ | Coefficient $\times 10^6/^{\circ}\text{F}$ | Scatter $\times 10^6/^{\circ}\text{F}$ |
| 400 | 2.10 | 0.36 | 1.38 | 0.59 | 0.77 | 0.55 |
| 600 | 2.14 | 0.56 | 1.38 | 0.42 | 0.94 | 0.39 |
| 800 | 2.09 | 0.56 | 1.40 | 0.29 | 1.09 | 0.36 |
| 1000 | 2.02 | 0.38 | 1.45 | 0.23 | 1.21 | 0.37 |
| 1200 | 1.99 | 0.31 | 1.49 | 0.28 | 1.32 | 0.38 |
| 1400 | 2.00 | 0.28 | 1.52 | 0.45 | 1.40 | 0.37 |
| 1600 | 2.05 | 0.32 | 1.57 | 0.42 | 1.47 | 0.38 |
| 1800 | 2.11 | 0.30 | 1.61 | 0.46 | 1.54 | 0.37 |
| 2000 | 2.14 | 0.27 | 1.66 | 0.35 | 1.57 | 0.34 |

The estimated standard deviation for sample 3 is probably the most reliable figure because it is based on 15 runs, whereas s values for samples 1 and 2 are based on 6 runs each. It is noteworthy that estimated standard deviations for samples 1 and 3 are practically the same.

In the evaluation of graphite data in subsequent sections the $2s$ level, i.e., $0.20 \times 10^{-6}/^{\circ}\text{F}$, is generally used. In questionable instances, i.e., those in which the deviation is very close to $2s$, additional experimental data were obtained to check the result.

The possibility that heating or reheating a graphite sample would produce a permanent change in its thermal expansion characteristics was examined. In Table 4 repetitive data are shown for three grades of molded and extruded graphite. In 3 of the 13 groups of runs the first mean coefficient of expansion appears to be considerably different from the other coefficients in the list. Specifically, the third sample of block 1101 graphite gave a coefficient of $1.95 \times 10^{-6}/^{\circ}\text{F}$ in the first run, whereas the coefficients for the five succeeding runs were considerably higher. Similarly, the fourth sample of block 1101 gave a coefficient of $1.47 \times 10^{-6}/^{\circ}\text{F}$ in the first run, and the coefficients for the five succeeding runs were much higher. Finally, the fourth sample of block 0104 gave a coefficient of $2.34 \times 10^{-6}/^{\circ}\text{F}$ in the first run, whereas the coefficients for the two succeeding runs were considerably lower. For the third and fourth samples of block 1101, estimated standard deviations based on all but the first

Table 4. Repetitive Determination of Linear Coefficients of Thermal Expansion Between 80 and 2000°F

| Sample Designation | Run | Mean Coefficient of Linear Thermal Expansion $\times 10^6/^{\circ}\text{F}$ (80 to 2000°F) |
|---|-----|---|
| Extruded graphite, block 1101, 1st sample ^a | 1 | 2.04 |
| | 2 | 1.64 |
| | 3 | 2.13 |
| Extruded graphite, block 1101, 2nd sample ^a | 1 | 2.29 |
| | 2 | 2.32 |
| Extruded graphite, block 1101, 3rd sample ^a | 1 | 1.95 |
| | 2 | 2.16 |
| | 3 | 2.20 |
| | 4 | 2.21 |
| | 5 | 2.22 |
| | 6 | 2.12 |
| Extruded graphite, block 1101, 4th sample ^a | 1 | 1.47 |
| | 2 | 1.72 |
| | 3 | 1.64 |
| | 4 | 1.65 |
| | 5 | 1.82 |
| | 6 | 1.70 |
| Extruded graphite, block 1101, 5th sample ^a | 1 | 1.66 |
| | 2 | 1.54 |
| | 3 | 1.69 |
| | 4 | 1.54 |
| | 5 | 1.56 |
| | 6 | 1.58 |
| | 7 | 1.67 |
| | 8 | 1.38 |
| | 9 | 1.69 |
| | 10 | 1.56 |
| | 11 | 1.68 |
| | 12 | 1.60 |

^aParallel to extrusion axis.

Table 4 (Cont'd)

| Sample Designation | Run | Mean Coefficient of Linear Thermal Expansion $\times 10^6/^{\circ}\text{F}$ (80 to 2000 $^{\circ}\text{F}$) |
|---|-----|--|
| Extruded graphite, block 1101, 5th sample ^a (cont'd) | 13 | 1.54 |
| | 14 | 1.35 |
| | 15 | 1.58 |
| Extruded graphite, block 0301 ^a | 1 | 1.89 |
| | 2 | 1.82 |
| Molded graphite, block 0105, 1st sample ^b | 1 | 2.57 |
| | 2 | 2.52 |
| | 3 | 2.54 |
| | 4 | 2.56 |
| | 5 | 2.39 |
| Molded graphite, block 0104, 2nd sample ^c | 6 | 2.61 |
| | 1 | 1.99 |
| | 2 | 2.07 |
| | 3 | 1.94 |
| | 4 | 2.02 |
| Molded graphite, block 0104, 3rd sample ^c | 5 | 1.98 |
| | 6 | 2.13 |
| | 1 | 1.98 |
| | 2 | 2.00 |
| | 3 | 1.93 |
| Molded graphite, block 0104, 4th sample ^c | 4 | 2.02 |
| | 5 | 2.01 |
| | 6 | 2.02 |
| Molded graphite, block 0104, 5th sample ^c | 1 | 2.34 |
| | 2 | 1.93 |
| | 3 | 1.97 |
| Molded graphite, block 0104, 6th sample ^c | 1 | 2.01 |
| | 2 | 1.96 |
| | 3 | 1.96 |
| Molded graphite, block 0104, 7th sample ^c | 1 | 2.03 |
| | 2 | 2.03 |
| Molded graphite, block 0104, 8th sample ^c | 1 | 2.01 |
| | 2 | 1.98 |

^aParallel to extrusion axis.
^bParallel to molding direction.
^cPerpendicular to molding direction.

coefficients were as follows: third sample, $0.041 \times 10^{-6}/^{\circ}\text{F}$; fourth sample, $0.072 \times 10^{-6}/^{\circ}\text{F}$.

In the third sample, the first coefficient differs from the group average of the remaining coefficients by $0.23 \times 10^{-6}/^{\circ}\text{F}$, an amount greater than 5 times the estimated standard deviation. In a normal distribution this degree of departure would undoubtedly be considered significant. Experience has shown that in dilatometry a long negative "tail" is frequently found in the distribution curve. This is illustrated by the eighth and fourteenth coefficients for the fifth sample of block 1101. The evidence from the third sample is therefore not conclusive. In the fourth sample of block 1101, the first coefficient differs from the group average of the remaining coefficients by $0.24 \times 10^{-6}/^{\circ}\text{F}$, slightly more than 3 times the

estimated standard deviation. The data on the fourth sample of block 0104 are too scanty to permit statistical treatment.

Apart from the three instances discussed above, no consistent alteration in thermal expansion coefficient can be inferred. The variations recorded tend toward randomness and are attributed to the method and equipment. Every material has been rerun a sufficient number of times to prevent the inclusion of abnormally high or low results.

It may be further noted that standard deviations computed for 6 runs on electrolytic copper and 10 runs on "A" nickel were somewhat larger than any of the corresponding standard deviations computed for block 1101 extruded graphite. For the 80 to 1400 $^{\circ}\text{F}$ temperature interval the estimated standard deviations were as follows:

electrolytic copper: $0.20 \times 10^{-6}/^{\circ}\text{F}$

"A" nickel: $0.17 \times 10^{-6}/^{\circ}\text{F}$

block 1101 graphite: $0.11 \times 10^{-6}/^{\circ}\text{F}$

B. Mean Linear Coefficients of Thermal Expansion

Percentage expansion vs temperature curves for blocks 0104 (molded) and 0201 (extruded) in the three mutually perpendicular directions are plotted in Figs. 3 and 4. These curves are characterized by slopes which gradually increase with temperature. The curves in Figs. 3 and 4 are representative of all graphites in that they show a greater thermal expansion perpendicular to the grain direction than parallel to it.

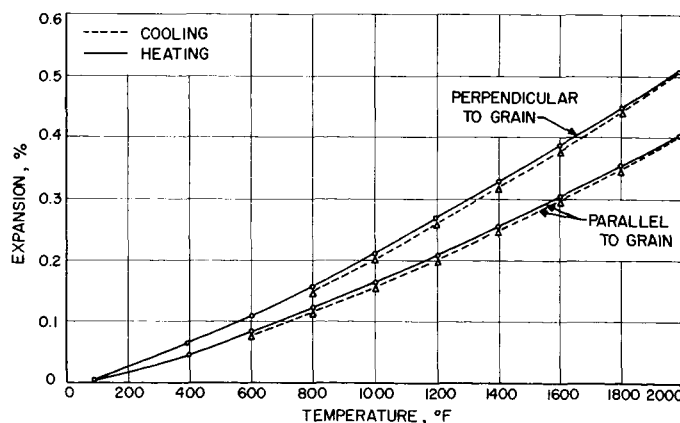


Fig. 3. Total Linear Expansion vs Temperature, Block 0104 (Molded)

Curves of mean linear coefficient of thermal expansion vs temperature for two blocks are shown in Figs. 5 and 6. The mean linear coefficients of thermal expansion for all blocks tested are given in Table 5.

C. Mean Cubical Coefficients of Thermal Expansion

The mean coefficients of cubical thermal expansion are given in Table 6. The cubical coefficient is very nearly equal to the sum of the three mutually perpendicular linear coefficients. The exact cubical coefficient is given by the equation

$$\begin{aligned} \text{cubical coefficient} &= (1 + a)(1 + b)(1 + c) - 1 \\ &= a + b + c + ab + bc + ac + abc \end{aligned}$$

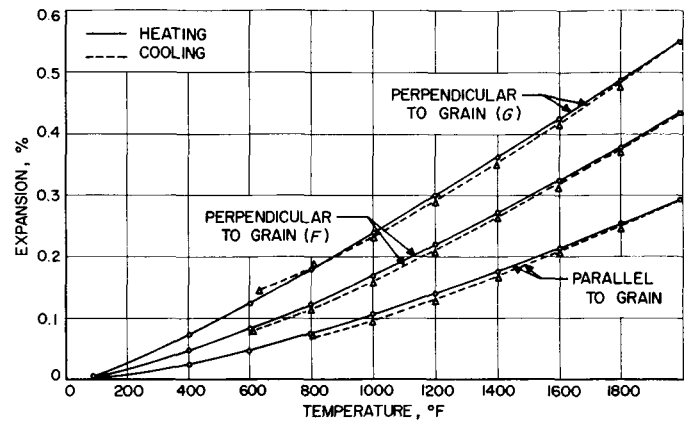


Fig. 4. Total Linear Expansion vs Temperature, Block 0201 (Extruded)

Table 5. Mean Linear Coefficients of Thermal Expansion of Graphites

| Block and Orientation | Coefficient of Expansion $\times 10^6 / ^\circ\text{F}$ from 80°F to | | | | | | | | |
|-----------------------|--|---------------------|---------------------|----------------------|----------------------|----------------------|----------------------|----------------------|----------------------|
| | 400°F | 600°F | 800°F | 1000°F | 1200°F | 1400°F | 1600°F | 1800°F | 2000°F |
| Molded | | | | | | | | | |
| 0104 M | 1.85 | 2.06 | 2.17 | 2.28 | 2.37 | 2.45 | 2.55 | 2.61 | 2.64 |
| 0104 N | 1.59 | 1.62 | 1.69 | 1.77 | 1.84 | 1.92 | 2.00 | 2.05 | 2.10 |
| 0104 P | 1.31 | 1.43 | 1.56 | 1.67 | 1.77 | 1.85 | 1.94 | 2.01 | 2.08 |
| 0105 M | 1.68 | 1.92 | 2.14 | 2.30 | 2.46 | 2.50 | 2.57 | 2.61 | 2.65 |
| 0105 N | 1.17 | 1.35 | 1.50 | 1.58 | 1.71 | 1.78 | 1.87 | 1.94 | 1.98 |
| 0105 P | 1.43 | 1.55 | 1.69 | 1.84 | 1.85 | 1.95 | 2.02 | 2.09 | 2.13 |
| 0107 M | 1.47 | 1.74 | 1.93 | 2.08 | 2.22 | 2.34 | 2.42 | 2.51 | 2.56 |
| 0107 N | 1.27 | 1.49 | 1.62 | 1.76 | 1.85 | 1.91 | 2.00 | 2.09 | 2.13 |
| 0107 P | 1.47 | 1.55 | 1.67 | 1.79 | 1.89 | 1.95 | 2.00 | 2.09 | 2.13 |
| 0501 M | 1.76 | 1.88 | 1.98 | 2.08 | 2.16 | 2.23 | 2.32 | 2.37 | 2.41 |
| 0501 N | 1.41 | 1.55 | 1.61 | 1.71 | 1.78 | 1.89 | 1.94 | 2.01 | 2.04 |
| 0501 P | 1.40 | 1.50 | 1.66 | 1.74 | 1.80 | 1.89 | 1.93 | 1.98 | 2.02 |
| 0601 M | 1.96 | 2.15 | 2.27 | 2.40 | 2.43 | 2.50 | 2.54 | 2.56 | 2.52 |
| 0601 N | 1.69 | 1.72 | 1.84 | 1.88 | 1.96 | 2.05 | 2.15 | 2.17 | 2.22 |
| 0601 P | 1.74 | 1.79 | 1.88 | 1.98 | 2.07 | 2.13 | 2.23 | 2.30 | 2.34 |
| 0801 M | 2.02 | 2.18 | 2.30 | 2.34 | 2.42 | 2.48 | 2.54 | 2.59 | 2.65 |
| 0801 N | 1.51 | 1.60 | 1.66 | 1.77 | 1.83 | 1.90 | 1.95 | 2.00 | 2.00 |
| 1001 M | 2.40 | 2.60 | 2.79 | 2.87 | 2.94 | 3.03 | 3.09 | 3.16 | 3.15 |
| 1001 N | 1.66 | 1.69 | 1.74 | 1.79 | 1.87 | 1.93 | 1.97 | 2.03 | 2.02 |
| 1001 P | 1.12 | 1.26 | 1.39 | 1.47 | 1.58 | 1.69 | 1.77 | 1.84 | 1.89 |
| 1002 M | 3.06 | 3.09 | 3.13 | 3.17 | 3.24 | 3.34 | 3.37 | 3.43 | 3.45 |
| 1002 N | 0.84 | 1.14 | 1.30 | 1.41 | 1.53 | 1.58 | 1.66 | 1.73 | 1.78 |
| 1002 P | 1.04 | 1.09 | 1.24 | 1.39 | 1.48 | 1.56 | 1.64 | 1.70 | 1.75 |
| Extruded | | | | | | | | | |
| 0201 E | 0.77 | 0.90 | 1.03 | 1.12 | 1.25 | 1.33 | 1.41 | 1.45 | 1.50 |
| 0201 F | 1.39 | 1.57 | 1.70 | 1.83 | 1.96 | 2.05 | 2.14 | 2.18 | 2.26 |
| 0201 G | 2.37 | 2.44 | 2.43 | 2.62 | 2.67 | 2.73 | 2.78 | 2.83 | 2.85 |
| 0301 E | 1.01 | 1.19 | 1.33 | 1.44 | 1.57 | 1.64 | 1.71 | 1.79 | 1.82 |
| 0301 F | 1.83 | 1.92 | 2.06 | 2.12 | 2.24 | 2.31 | 2.35 | 2.44 | 2.53 |
| 0301 G | 1.51 | 1.69 | 1.81 | 1.95 | 1.96 | 2.06 | 2.16 | 2.25 | 2.36 |
| 0401 E | 1.55 | 1.72 | 1.89 | 1.94 | 2.07 | 2.13 | 2.23 | 2.27 | 2.32 |
| 0401 F | 2.46 | 2.55 | 2.66 | 2.74 | 2.81 | 2.91 | 2.96 | 3.02 | 3.06 |
| 0401 G | 2.39 | 2.58 | 2.67 | 2.75 | 2.85 | 2.92 | 2.99 | 3.08 | 3.10 |
| 0701 E | 1.28 | 1.35 | 1.32 | 1.39 | 1.47 | 1.53 | 1.59 | 1.63 | 1.63 |
| 0701 G | 1.50 | 1.85 | 1.92 | 2.07 | 2.18 | 2.24 | 2.34 | 2.41 | 2.43 |

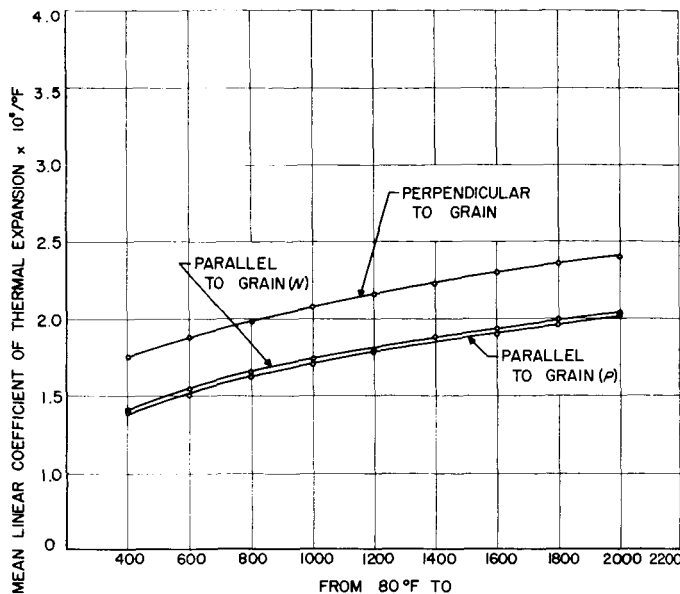


Fig. 5. Mean Linear Coefficients of Thermal Expansion vs Temperature, Block 0501 (Molded)

where a , b , and c are the linear coefficients. The last four terms are very small relative to the first three and may be dropped.

D. Effect of Location and Orientation

Eighteen specimens were prepared from a $12 \times 12 \times 2\frac{1}{4}$ -in. block of fine-grain molded graphite; these were tested for variations in coefficient of thermal expansion with respect to (1) location in the block and (2) orientation in the block. Figure 7 shows the outcome of these experiments. The rectangular parallelepipeds represent the pieces initially cut from the block and from which the cylindrical thermal expansion specimens were machined.

The ranges of variation of coefficients for the three directions of sample orientation, as given in Fig. 1, are as follows: $M-0.07$, $N-0.19$, and $P-0.40 \times 10^{-6}/^{\circ}\text{F}$. None of these ranges exceeds the limits for the $2s$ confidence level, although the range of P -direction samples is equal to the $2s$ range because of the coefficient determined for sample 17. Two subsequent runs on sample 17 gave substantially lower figures, i.e., 1.93 and $1.97 \times 10^{-6}/^{\circ}\text{F}$; therefore, this sample is not considered to have an abnormally high coefficient.

These results would indicate that thermal expansion is not dependent on location in the block; one sample taken

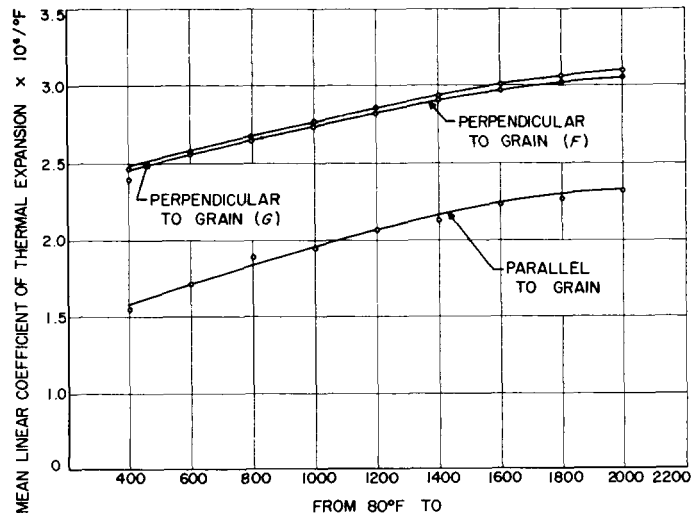


Fig. 6. Mean Linear Coefficients of Thermal Expansion vs Temperature, Block 0401 (Extruded)

from a random location should be sufficient for the measurement of the coefficient of thermal expansion in a given direction in a molded block of comparable size and uniformity.

Another purpose of the preceding series of experiments was to compare the coefficients of expansion for the two directions perpendicular to the molding pressure (N and P), irrespective of the location within the block. The average coefficients of expansion for these directions are:

$$N\text{-direction: } 2.091 \times 10^{-6}/^{\circ}\text{F}$$

$$P\text{-direction: } 2.093 \times 10^{-6}/^{\circ}\text{F}$$

Application of the statistical t -test yields overwhelming odds in favor of the hypothesis that these two averages are statistically identical. (This supports the conclusion that the apparently high coefficient first measured on sample 17 is not significant.) Thus, the maximum and minimum coefficients of thermal expansion of a block of molded graphite can be determined from two samples, one parallel and one perpendicular to the molding direction.

E. Relationship Between Thermal Expansion and Electrical Resistivity

Electrical resistivities at 32°F for molded and extruded graphite in the three mutually perpendicular directions are presented in Table 7. These resistivities are plotted

Table 6. Mean Cubical Coefficients of Thermal Expansion of Graphites

| Block | Coefficient of Cubical Expansion $\times 10^6 / ^\circ\text{F}$ from 80°F to | | | | | | | | |
|-------------------|--|-------|-------|--------|--------|--------|--------|--------|--------|
| | 400°F | 600°F | 800°F | 1000°F | 1200°F | 1400°F | 1600°F | 1800°F | 2000°F |
| Molded | | | | | | | | | |
| 0104 | 4.75 | 5.11 | 5.42 | 5.72 | 5.98 | 6.22 | 6.49 | 6.67 | 6.82 |
| 0105 | 4.28 | 4.82 | 5.33 | 5.72 | 6.02 | 6.23 | 6.46 | 6.64 | 6.78 |
| 0107 | 4.01 | 4.78 | 5.22 | 5.63 | 5.96 | 6.20 | 6.42 | 6.69 | 6.82 |
| 0501 | 4.57 | 4.93 | 5.25 | 5.53 | 5.74 | 6.01 | 6.19 | 6.36 | 6.47 |
| 0601 | 5.39 | 5.66 | 5.99 | 6.26 | 6.46 | 6.68 | 6.92 | 7.03 | 7.08 |
| 0801 ^a | 5.04 | 5.38 | 5.62 | 5.88 | 6.08 | 6.28 | 6.44 | 6.59 | 6.65 |
| 1001 | 5.18 | 5.55 | 5.92 | 6.13 | 6.39 | 6.65 | 6.83 | 7.03 | 7.06 |
| 1002 | 4.94 | 5.32 | 5.67 | 5.97 | 6.25 | 6.48 | 6.67 | 6.86 | 6.98 |
| Extruded | | | | | | | | | |
| 0201 | 3.55 | 4.04 | 4.43 | 4.78 | 5.17 | 5.43 | 5.69 | 5.81 | 6.02 |
| 0301 | 4.35 | 4.80 | 5.20 | 5.51 | 5.77 | 6.01 | 6.22 | 6.48 | 6.71 |
| 0401 | 6.40 | 6.85 | 7.22 | 7.43 | 7.73 | 7.96 | 8.18 | 8.37 | 8.48 |
| 0701 ^b | 4.28 | 5.05 | 5.16 | 5.53 | 5.83 | 6.01 | 6.27 | 6.45 | 6.49 |

^aCoefficient of linear expansion in the M-direction plus 2 times the coefficient of linear expansion in the N-direction. The P-direction was not available in sufficiently long pieces.

^bCoefficient of linear expansion in the E-direction plus 2 times the coefficient of linear expansion in the G-direction. The F-direction was not available in sufficiently long pieces.

Table 7. Electrical Resistivities of Graphites

| Block and Orientation | Electrical Resistivity $\times 10^3$ ohm cm at 32°F |
|-----------------------|--|
| Molded | |
| 0104 M | 1.390 |
| 0104 N | 1.084 |
| 0104 P | 1.128 |
| 0105 M | 1.351 |
| 0105 N | 1.076 |
| 0105 P | 1.031 |
| 0107 M | 1.356 |
| 0107 N | 1.036 |
| 0107 P | 1.059 |
| 0501 M | 1.318 |
| 0501 N | 1.066 |
| 0501 P | 1.092 |
| 0601 M | 1.765 |
| 0601 N | 1.462 |
| 0601 P | 1.471 |
| 0801 M | 1.719 |
| 0801 N | 1.200 |
| 1001 M | 1.998 |

| Block and Orientation | Electrical Resistivity $\times 10^3$ ohm cm at 32°F |
|-----------------------|--|
| Molded | |
| 1001 N | 1.354 |
| 1001 P | 1.128 |
| 1002 M | 1.948 |
| 1002 N | 1.071 |
| 1002 P | 1.058 |
| Extruded | |
| 0201 E | 0.7496 |
| 0201 G | 1.305 |
| 0301 E | 0.9325 |
| 0301 F | 1.203 |
| 0301 G | 1.139 |
| 0401 E | 1.336 |
| 0401 F | 1.673 |
| 0401 G | 1.631 |
| 0701 E | 0.932 |
| 0701 G | 1.508 |

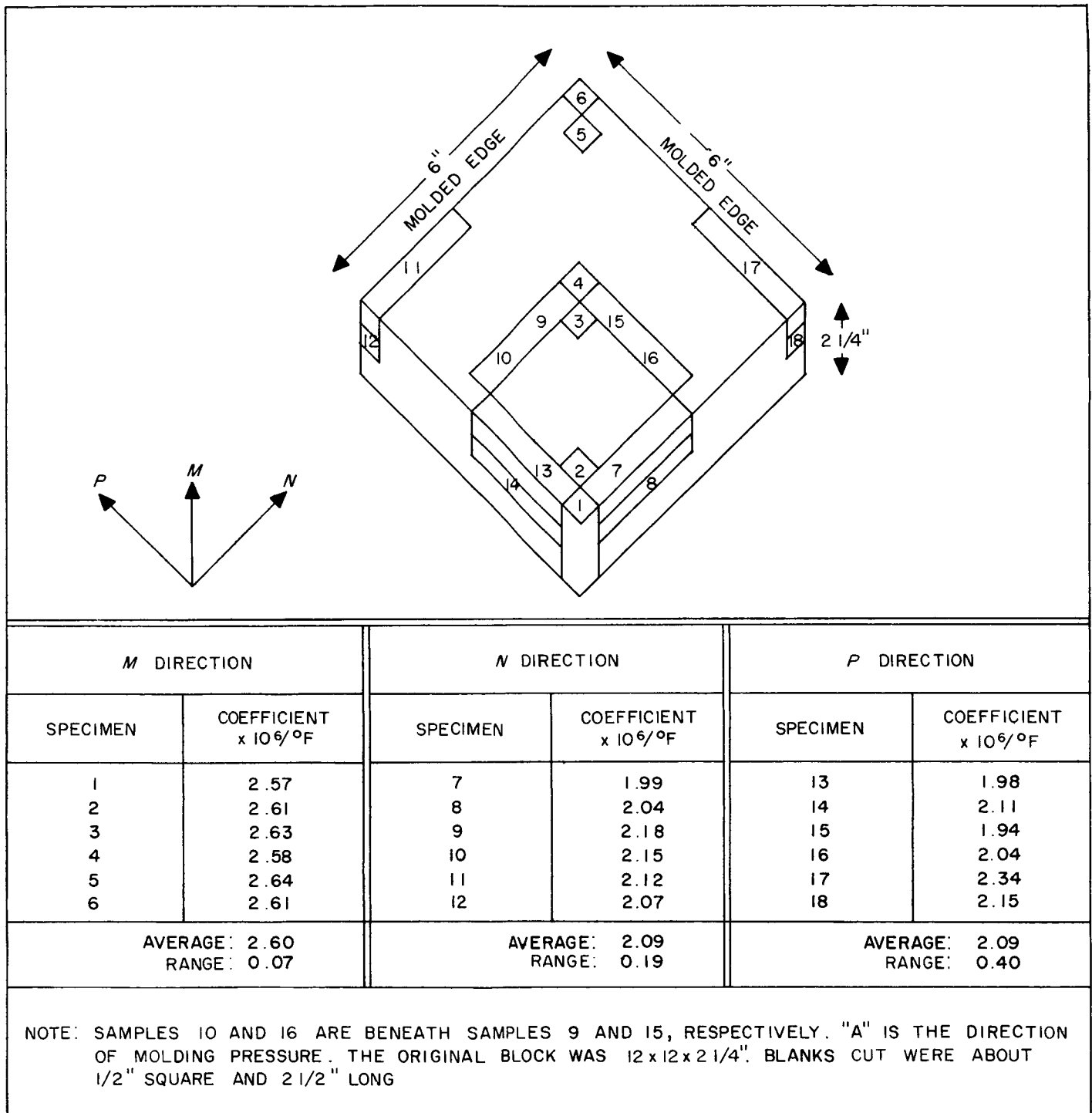


Fig. 7. Locations, Orientations, and Mean Linear Coefficients of Linear Thermal Expansion (80–2000°F) of Specimens Taken from Block 0104 (Molded)

against the corresponding mean linear coefficients of thermal expansion (80 to 2000°F) in Fig. 8.

It is readily obvious from these data that a tendency toward proportionality exists; i.e., low coefficients of thermal expansion tend to be associated with low resistivities and vice versa. With regard to crystallite orientation, it is reasonable to expect a higher resistivity perpendicular to an individual crystal plate than parallel to it.

The resistivities of nine samples were measured before and after thermal expansion runs for variations due to heating to 2000°F. In most cases the differences between these resistivities were less than twice the standard deviation of the mean.

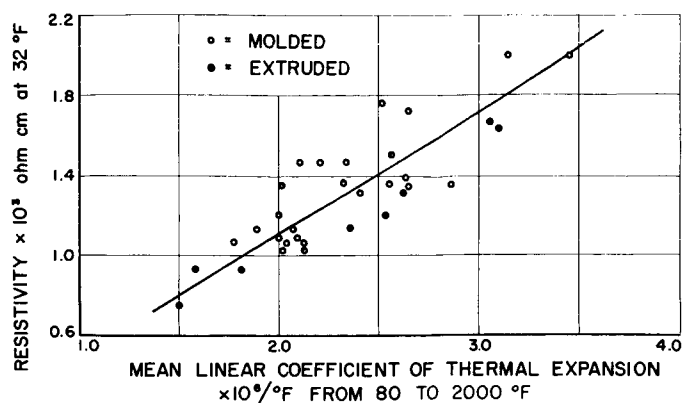


Fig. 8. Resistivity (at 32°F) vs Mean Linear Coefficient of Thermal Expansion (80–2000°F) of Molded and Extruded Graphites

V. DISCUSSION OF RESULTS

In Table 5, the molded graphites may be conveniently divided into two groups: (a) 0104, 0105, 0107, 0501, 0601, and 0801; (b) 1001 and 1002. Note that samples in group (b) were taken from the same grade. The principal difference between these groups is that the directional anisotropy of mean linear coefficients of thermal expansion shown by the two blocks in group (b) is significantly greater than that shown by the six graphites in group (a). For group (b) the coefficients in the molding direction are greater and the coefficients in the grain directions generally smaller than the corresponding coefficients for group (a). The larger anisotropy of group (b) may be attributed to the fact that this grade was baked under pressure. This grade also gave a larger than average anisotropy with respect to electrical resistivity. The fact that this grade was not graphitized may also bear on these results.

Considering the individual materials within group (a) it is apparent from Table 5 that the experimental differences recorded for each of the three directions are relatively small. The maximum differences are as follows:

$$M\text{-direction: } 0.24 \times 10^{-6}/^{\circ}\text{F}$$

$$N\text{-direction: } 0.24 \times 10^{-6}/^{\circ}\text{F}$$

$$P\text{-direction: } 0.32 \times 10^{-6}/^{\circ}\text{F}$$

The largest spread is $\pm 0.16 \times 10^{-6}/^{\circ}\text{F}$, less than twice the standard deviation. The directional anisotropy is least for block 0601; if significant, this result may be due to its manufacture with graphite flour instead of coke. Block 0501 was manufactured according to the same specifications as blocks 0104, 0105, and 0107. Block 0801 was made from the same initial ingredients but by a different manufacturer. Although block 0601 was made from a graphite flour instead of coke, its coefficient of expansion was not found to express this difference.

It is important to mention that the linear coefficients for the two grain directions (*N* and *P*) in any one of the above-described molded graphites lie within reasonable experimental error of one another.

Referring to Table 6, the coefficients of cubical thermal expansion for all eight molded graphites exhibit a range of $0.61 \times 10^{-6}/^{\circ}\text{F}$. Since the cubical coefficient consists of the sum of three mutually perpendicular linear coefficients, the range shown by a group of cubical coefficients

could be as great as the sum of the three linear ranges found for the same group. The above figure is therefore not considered significant.

From Table 5, it is apparent that the respective linear coefficients found for block 0401 are significantly higher than those found for the other three. This observation is of interest since this grade was not completely graphitized. Its electrical resistivities are high, relative to those of the other extruded graphites. If blocks 0201, 0301, and 0701 are considered as a group, the ranges are:

$$E\text{-direction: } 0.32 \times 10^{-6}/^{\circ}\text{F}$$

$$F\text{-direction: } 0.27 \times 10^{-6}/^{\circ}\text{F}$$

$$G\text{-direction: } 0.49 \times 10^{-6}/^{\circ}\text{F}$$

The ranges for *E* and *F* directions are about $1\frac{1}{2}$ times the standard deviation (plus and minus), and the range for the *G* direction is about $2\frac{1}{2}$ times the standard deviation (plus and minus). Among these three no significant difference is established.

In block 0201 the coefficient for the *F* direction is considerably less than that for the *G* direction (Fig. 4). Several repeat runs on new samples with these directions also showed a sizable difference between their coefficients. This is the only grade of graphite considered in this study in which a triaxiality of thermal expansion characteristics was indicated. This phenomenon may be attributed to the fact that this material was extruded with a rectangular cross section.

The data obtained for the extruded graphites suggest that the degree of anisotropy is inversely related to the maximum filler particle size. The anisotropy shown by block 0301, the coarsest extruded graphite, is less than that shown by blocks 0201 and 0701, the finest extruded graphites.

Reimpregnation with pitch apparently has little if any effect on the coefficient of expansion. (Refer to blocks 0201 and 0701 in Tables 1 and 5.)

The mean coefficients of cubical thermal expansion for extruded graphite (Table 6) reflect the findings described above. The cubical coefficients for blocks 0201, 0301, and 0701 show a range of $0.69 \times 10^{-6}/^{\circ}\text{F}$, slightly larger than the range reported for molded graphites. The range is

larger probably because of a wider spread in grain sizes among the extruded graphites. The cubical coefficient for block 0401 is larger, reflecting the larger linear coefficients previously discussed.

When all the cubical coefficients for both molded and extruded graphite are considered, it is seen that block 0401 has the largest and block 0201 the smallest.

If the mean linear coefficients of thermal expansion of Table 5 are plotted against temperature interval it is found that the slopes of the curves for a given temperature interval do not differ widely from one grade to another or from one direction to another. This observation was made by Currie, Hamister, and MacPherson (Ref. 3); assuming curve parallelism, they suggested the determination of coefficients of expansion up to 2500°C by extrapolation from the measured coefficient over the temperature interval from 20 to 100°C. The data gathered in the present investigation have been utilized to check the relative validity of this procedure for these materials. Figure 9 shows the comparison between "values to be added," published by Currie et al, and those calculated from the present thermal expansion data which showed the largest deviations from the Currie data.

To examine the significance of this comparison, assume a mean linear coefficient of thermal expansion of

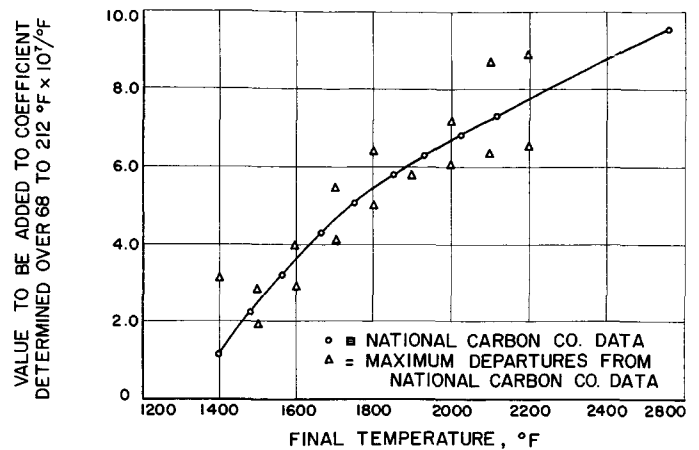


Fig. 9. Value To Be Added to Coefficient (68–212°F) vs Temperature

$1.50 \times 10^{-6}/^{\circ}\text{F}$ for the interval from room temperature to 212°F. From Fig. 9 the maximum variation in "value to be added" to obtain the coefficient for the interval room temperature to 2000°F is between 0.65 and $0.89 \times 10^{-6}/^{\circ}\text{F}$. The extreme coefficients which can be calculated are 2.15 and $2.39 \times 10^{-6}/^{\circ}\text{F}$. Thus, the maximum scatter is near $\pm 5\%$. The value suggested by Currie et al is $0.775 \times 10^{-6}/^{\circ}\text{F}$, almost midway between the extremes. Finally, these experimental data support the extrapolative method of determining mean linear coefficient of thermal expansion.

VI. CONCLUSIONS

From the results of the investigation the following conclusions are drawn:

1. The mean linear coefficient of thermal expansion of graphite increases gradually with temperature. The coefficients for various samples and orientations show approximately the same temperature dependence; i.e., curves of mean coefficient vs temperature interval have comparable slopes.
2. The Currie-Hamister-MacPherson method for determining the coefficient of thermal expansion is substantiated.
3. Specimens cut perpendicular to the extrusion direction or parallel to the molding direction have greater expansions than specimens cut parallel to the extrusion direction or perpendicular to the molding direction.
4. Among three commercial molded grades, no proven differences in thermal expansion characteristics were found in any given direction. The coefficients of expansion for the two directions perpendicular to the molding direction are within reasonable experimental error of one another.
5. Thermal expansion is not demonstrably dependent on location within a molded block; one sample taken from a random location is sufficient for the measurement of its coefficient in a given direction. Two mutually perpendicular directions which are both perpendicular to the molding direction are shown to be statistically identical with respect to thermal expansion.
6. In molded graphite the use of graphite flour instead of petroleum coke may reduce the directional anisotropy of expansion.
7. Molded carbonaceous material baked under pressure appears to have a larger directional anisotropy of expansion than molded graphite produced by conventional baking.
8. Graphite extruded through a rectangular opening shows triaxial anisotropy with respect to thermal expansion.
9. Incomplete graphitization may increase the coefficient of expansion.
10. In extruded graphite the degree of anisotropy of the coefficient of expansion may be inversely related to the maximum filler particle size.
11. The coefficient of expansion of graphite from 80 to 2000°F is not altered by repeated heating and cooling of the material in this temperature interval.
12. Electrical resistivity (at 32°F) and mean linear coefficient of thermal expansion (80 to 2000°F) show a pronounced tendency toward proportionality.

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